IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

MOK ET AL

Serial No.: 09/361,626

Filed: July 27, 1999

Title: PROCESS FOR REMOVING ALUMINUM SPECIES FROM ALKALI METAL HALIDE BRINE SOLUTIONS

Group Art Unit: 1724

Examiner: Cintins

June 30, 2003

APPELLANTS' APPEAL BRIEF

Mail Stop Appeal Brief - Patents
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Sir:

Introduction

The invention on appeal is a method for reducing the content of soluble aluminum species in an evaporated salt alkali metal halide brine so as to provide a brine feedstock suitable for use in a chlor-alkali membrane cell process.

Real Party in Interest

The real party in interest is Kvaerner Canada Inc. by virtue of assignment recorded on October 5, 1999 on Reel 010292, Frame 0891.

Related Appeals and Interferences

There are no other appeals or interferences known to the appellants, the appellants' legal representative or assignee which will directly affect or be directly affected by, or have a bearing on, the Board's decision in the present appeal.

Status of Claims

Claims 1-7, 9 and 10 are pending and at issue in this appeal. Claim 8, the only other claim presented, has been canceled.

The claims at issue are set out in the Appendix to this brief.

Status of Amendments

No amendments were filed after the final rejection.

Summary of the Invention

As indicated, the invention at issue is concerned with reducing the amount of soluble aluminum species in an alkali metal brine so as to provide a brine feedstock which can be effectively used in a chlor-alkali membrane cell process. It is well known that the electrical efficiency of membrane cells is undesirably affected by the presence of anionic or cationic impurities, such as aluminum, and a great deal of effort has been directed towards removing aluminum species from brine which is to be used in chlor-alkali plants equipped with membrane cells. See pages 1-5 of appellants' specification.

Despite these efforts, however, significant problems persist in the effective removal of aluminum species from brine solutions which are to be used in chlor-alkali membrane cells. Page 5, lines 10-11 of appellants' specification.

The invention provides a highly useful method for the efficient removal of aluminum species from brine. According to the invention, this is accomplished by treating the brine with a magnesium salt in an amount sufficient to provide a Mg/Al molar ratio of 5-20 to 1 and a Mg concentration of from 0.5 to 5 ppm. Additionally, sufficient alkali metal hydroxide is added to provide an excess alkalinity concentration of between 0.1 and 0.3 grams per liter alkali metal hydroxide. This precipitates a magnesium aluminum hydroxide complex, the latter being removed to provide the desired brine feedstock. See appellants' specification, page 5, last full ¶ and last ¶, page 6 as well as claim 1.

Thus, by using the indicated combination of conditions, i.e. Mg/Al molar ratio 5-20 to 1, Mg concentration of 0.5 to 5 ppm and excess alkalinity concentration of between 0.1-0.3 g/L alkali metal hydroxide so as to precipitate a magnesium aluminum hydroxide

complex, it is possible to reduce the aluminum content so that the thus-treated brine can be effectively used in a chlor-alkali membrane cell without prior art problems.

Preferably the Mg/Al molar ratio is about 10:1, the Mg concentration is from 1 to 5 ppm and the excess alkalinity is 0.1-0.2 g/L alkali metal hydroxide. See last ¶, page 6 of appellants' specification and claims 2 and 7.

The advantages in aluminum removal efficiency using the combination of conditions called for by the appellants is shown in Table 2, page 12 of the specification. Compare, for example, the 91% removal efficiency for the first entry in Table 2, which is according to the invention, with the fifth entry where the efficiency drops to 72% when the excess alkalinity is outside the applicants' range.

Grouping of Claims

The Examiner has grouped all of the claims (claims 1-7, 9 and 10) together in rejecting the claims. The appellants consider that the issues can be fairly considered with all of the claims groups together.

The Issue

The Examiner has finally rejected all of the claims under 35 U.S.C. 103(a) as unpatentable over Nagy U.S. Patent 4,073,706. The patent is discussed in the paragraph bridging pages 4-5 of appellants' specification. As there discussed, the appellants note that Nagy states that less than 5 ppm of magnesium concentration is ineffective for aluminum removal from a brine solution and that Nagy obtains no removal of aluminum from a brine solution containing 1 ppm aluminum and only 2.5 ppm magnesium (i.e. below the appellants' required molar ratio of 5-20 to 1) and 0.1 g/l NaOH after 20 minutes. It is believed that this in and of itself shows unobviousness in the appellants' invention.

In any case, based on the Examiner's rejection, the main issue for consideration herein is:

> whether or not the Examiner has erred in considering the appellants' invention to be obvious from Nagy.

A more specific issue is whether or not declaration evidence submitted by the appellants has established unobviousness in the appellants' invention over the Nagy disclosure.

The appellants submit that the issues should be decided in their favor, i.e. the appellants' invention is not obvious from Nagy and the declaration evidence of record confirms unobviousness in the appellants' invention. Accordingly, the appellants submit that the Examiner's Section 103(a) rejection should be reversed.

Argument

Before discussing the issues, it is believed useful to consider the Nagy disclosure and the appellants' declaration evidence of record (declaration of Dr. Zbigniew Twardowski, the R and D manager for the appellants' assignee).

Nagy U.S. Patent 4,073,706

Nagy is concerned with the removal of trace metals, e.g. aluminum, from brine by the addition of a magnesium compound to the brine to obtain a magnesium concentration of greater than 5 ppm. The magnesium is then precipitated as $Mg(OH)_2$ to remove contaminants (see Col. 1, 1st ¶; claim 1). At Column 2, line 37, Nagy indicates that soluble magnesium is added to the brine being treated to obtain a level of "at least about" 5 ppm Mg in the brine. The patentee specifically notes that levels of Mg "less than about 5 ppm" have been found to be ineffective in removing impurities (see Col. 2, lines 43-47). He suggests that much higher levels of Mg may be helpful and he proposes a range of about 5 to about 30 ppm Mg (Col. 2, lines 47-52). Patent claim 1 specifies adding magnesium to the brine to obtain an Mg concentration of greater than 5 ppm.

While the lowest limit (5 ppm Mg) mentioned by Nagy touches the appellants' upper limit, it is evident that Nagy's teaching is, for all intents and purposes, contrary to the appellants' invention where a magnesium salt is added to the brine to give an Mg concentration of from 0.5 to 5 ppm Mg concentration in combination with an Mg/Al molar ratio of 5-20, preferably about 10 to 1, and an excess alkalinity of 0.1 to 0.3 g/L alkali metal hydroxide. The unobviousness of the appellants' invention is highlighted by Example 2, Table II, of Nagy who shows 0% aluminum removal from a brine solution

containing 2.5 parts Mg and 1 part Al (i.e. well below the appellants' required molar ratio of 5-20 to 1 Mg to Al) and 0.1 g/L NaOH.

The Twardowski Declaration

This declaration has been presented in support of the unobviousness of the appellants' invention. It notes that the invention is based on the finding of a critical relationship in the Mg to Al molar ratio, Mg concentration and alkali metal hydroxide to provide excess alkalinity of 0.1 to 0.3 alkali metal hydroxide. The declaration includes the results of experiments (see Exhibit A thereof) which, in the appellants' view, convincingly establishes the unobviousness of the presently claimed invention. The test results given in Exhibit A are discussed in ¶s 12 and 13 of the declaration. Further test results are given in ¶ 14. Particular attention is called to Table 2, page 9 of the declaration which shows that a test using the appellants' combination of conditions, including an alkalinity excess of 0.1-0.2 g/l (see the last test in Table 2) gave a 99% Al removal while four other tests using Mg to Al molar ratios outside the appellants' claims gave significantly lower removal efficiencies.

Table 1, page 8 of the declaration, also shows high Al removal efficiencies while observing the appellants' conditions. The results are surprising, particularly when one considers Nagy's teaching against the use of less than 5 ppm Mg addition.

The declaration emphasizes the importance of the combination of conditions used, i.e. Mg and NaOH concentrations and Mg/Al molar ratios, and the unexpected results obtained thereof. See ¶ 16 of the declaration which summarizes the evidence.

Discussion of the Issues

In finally rejecting the appellants' claims, the Examiner has taken the position that Nagy discloses a process as claimed by the appellants except for the recited magnesium to aluminum molar ratio. However, the Examiner dismisses this ratio as not being seen to "materially affect the overall results of this process or to produce any new and unexpected result" (final rejection, page 2). Accordingly, he concludes that the magnesium to aluminum molar ratio is an obvious matter of choice.

The Examiner is in error in concentrating his position on patentability to the Mg/Al molar ratio called for by the appellants' claims. This ratio is indeed an important

difference over Nagy. However, even more significant is the fact that this feature needs to be used in combination with the recited Mg concentration and excess alkalinity recited in the claims. The appellants' invention is dependent on the combination of features, not just the Mg/Al molar ratio looked at in isolation. Nagy nowhere suggests the importance of this combination of features as required by appellants.

The Twardowski declaration shows the importance of the applicants' combination of conditions and the resulting unexpected results. The Examiner, however, dismisses the appellants' declaration evidence as not persuasive of patentability on the ground that it "fails to demonstrate new and unexpected results over the closest prior art" (final rejection, page 3). However, appellants submit that this is exactly what they have done. The Examiner states that the closest comparison to the Nagy process "appears to be Experiment # 1, run # 3, on page 8" of the Twardowski declaration. He notes that this run adds a magnesium salt to give a magnesium concentration of 5 ppm and also adds alkali metal hydroxide to provide excess alkalinity of 0.1-0.2 g/l. He also notes that the Mg to Al ratio in this run is 11:1. He then concludes (final rejection, bottom, page 3) that "since the Twardowski declaration fails to present any other test runs with different Mg/Al ratios at the same Mg concentration and excess alkalinity, the declaration does not demonstrate any new and unobvious results for the recited Mg to Al ratio over the closest prior art (i.e. Nagy)". However, this misses an important part of the test results given in Table 1. All three of the tests given in Table 1 are representative of the invention using variations within the appellants' claims to show these give high Al removal efficiency. The Examiner in an earlier action (Paper No. 20) took the view that the declaration showed unexpected results but not with features within the appellants' claims. See Paper No. 20. However, when it was explained to the Examiner that the declaration did more than that (see amendment of August 29, 2002), the Examiner has adopted the present view that the appellants have not, in essence, given a broader comparison against Nagy. However, the patentability of the appellants' invention is based on the showing that the specific combination of conditions called for by the appellants, is clearly new and gives results which are unexpected. The Examiner cannot validly deny this.

The Examiner goes on to say in the final rejection that the process of Nagy will inherently produce a brine having some magnesium to aluminum molar ratio and absent

a showing that the recited range produces a new and unexpected result, the exact magnesium to aluminum molar ratio employed by Nagy remains an obvious matter of choice, insufficient to patentably distinguish the claims.

The appellants do not challenge the fact that Nagy's method will have some Mg to Al ratio. This will happen when Mg is added to a brine containing aluminum to be removed. However, the fundamental deficiency of Nagy is that the reference does not suggest to one in the art that the appellants' combination of conditions, including the indicated Mg to Al molar ratio coupled with Mg concentration and specified excess alkalinity, gives unobvious efficiency in Al removal. This is the essence of appellants' invention. The declaration evidence shows the special advantages of using the specified combination of conditions. Nagy provides no suggestion of such a combination and, therefore, cannot make the appellants' invention obvious.

While the Twardowski declaration is self-explanatory in supporting the appellants' position as to unobviousness, the following summary as to the substance of the declaration is thought to be useful towards highlighting the appellants' position. Thus, in short, the declaration shows unobvious results using the following conditions representative of the applicants' invention:

Mg/Al molar ratio (claimed: 5-20/1, preferably about 10)

- 5.5 (Experiment No. 2, ¶ 14, page 9 of the declaration)
- 5.6 (Experiment Nos. 1 and 2 of declaration Exhibit A)
- 6.7 (Experiment No. 1, ¶ 14, page 8 of declaration)
- 8.9 (Experiment No. 1, ¶ 14, page 8 of declaration)
- 11.1 (Experiment Nos. 2, 3 of Exhibit A and Experiment No. 1, ¶ 14, page 8 of declaration)

Mg concentration (claimed: 0.5 to 5 ppm)

- 0.5 ppm (Experiment No. 1 of Exhibit A)
- 1.0 ppm (Experiment No. 2 of Exhibit A)
- 2.0 ppm (Experiment No. 3 of Exhibit A)
- 3-5 ppm (Experiment No. 1, ¶ 14, page 8 of declaration)

Alkalinity (claimed: 0.1 to 0.3 g/L, preferably 0.1 to 0.2 g/L)

0.1-0.2 g/L (Experiment Nos. 1, 2 and 3 of Exhibit A; Experiment Nos. 1 and 2, ¶ 14, pages 8-9 of declaration)

This showing is fairly and reasonably representative of the scope of the appellants' claims and, in the appellants' view, establishes unobviousness in the invention as claimed. Thus, Exhibit A of the declaration includes the results of three experiments, each of which includes nine separate runs or tests and a further experiment comprising eight different runs considered reasonably representative of Nagy. The various runs constituting Experiment Nos. 1-3 and the Experiment representative of Nagy were carried out under the same conditions except for the variations in the columns representing Al content in the starting brine, Mg added, Mg/Al molar ratio and NaOH. Removal efficiency was measured at the intervals (residence times) indicated.

The various tests were carried out by adding Mg and NaOH to brine containing Al at 50°C. This results in the precipitation of a complex of Mg, Al and OH which was removed, after which the resulting brines were checked for efficiency of Al removal. In Experiment Nos. 1-3, efficiency was measured immediately after addition of the Mg and NaOH and removal of the precipitate (i.e. zero residence time) and after 15 and 30 minutes residence times. The residence times for the Nagy experiment were 20 minutes, 40 minutes and 180 minutes at 66°C. The results of the experiments are tabulated in Exhibit A and graphically illustrated by the charts attached thereto. These results show how the efficiency of Al removal varies with Mg content, Mg/Al ratio and alkali concentration (NaOH).

The "Conclusion" section (pages 3-4 of Exhibit A) brings out the importance of observing the specific combination of conditions called for in the appellants' claims. This combination of conditions is not disclosed or suggested by Nagy (Diamond Shamrock) and the results thereof could not have been predicted from Nagy.

Of the Experiment No. 1 runs or tests, only the first two meet the requirements of the appellants' invention regarding Mg added, Mg/Al molar ratio and NaOH added. The conditions used which are representative of the appellants' invention are: 0.5 ppm Mg, 5.6 Mg/Al molar ratio and 0.1 or 0.2 NaOH g/L. The remaining runs of Experiment No. 1

use either more NaOH (the third run) and/or a lower Mg/Al molar ratio (all the other runs).

The conditions used in Experiment Nos. 2 and 3 representative of appellants' invention are 1.0 ppm Mg, 11.1 or 5.6 Mg/Al molar ratio and 0.1 or 0.2 g/L alkalinity (Experiment No. 2) or 2.0 ppm Mg, 11.1 Mg/Al molar ratio and 0.1 and 0.2 g/L alkalinity for Experiment No. 3.

As noted in the Twardowski declaration, the results of Experiment No. 3 are particularly striking as these show that runs representative of the invention (the 4th and 5th runs where the Mg/Al molar ratio is 11.1 and the NaOH content is 0.1 or 0.2) gave higher removal efficiencies than comparable runs (first and second) using double the Mg/Al ratio (22.2). While the removal efficiency for the first, second and third runs of Experiment No. 3 is good, the possibility of obtaining equivalent or, in this case, better removal efficiency at the lower Mg/Al ratio (11.1) consistent with the appellants' invention minimizes the possibility of introducing a further unwanted solids to the brine feedstock.

The Twardowski declaration (see ¶ 14) included additional test data confirming the unique results obtained by observing the specific conditions called for in the appellants' claims. Thus, Experiment No. 1 referred to in ¶ 14 of the declaration describes three separate experiments using 3 ppm, 4 ppm and 5 ppm of added Mg, Mg/Al molar ratios of 6.7, 8.9 and 11.1, respectively, and NaOH added to give an excess of between 0.1 and 0.2 g/L, again all within the appellants' claim 1. The test results show 98-99% aluminum removal efficiency.

Experiment No. 2, discussed beginning with the last ¶, page 8 of the Twardowski declaration, was similar to Experiment No. 1 except that the brine included 1 ppm aluminum rather than 0.5 ppm aluminum as in Experiment No. 1 and Mg/Al molar ratios were 1.1, 2.2, 3.3, 4.4 and 5.5, respectively. Only the test using the 5.5 Mg/Al molar ratio is within the appellants' invention.

The results of Experiment No. 2 as tabulated at page 9 of the declaration show the appellants' process giving an Al removal efficiency of 99% compared to significantly lower efficiencies using the Mg/Al molar ratios outside the appellants' claims.

It is respectfully submitted that the appellants' test data provides a fair comparison of representative conditions within the scope of the present claims and conditions just outside (note, for example, in the test results given in Table 2 of the

Twardowski declaration using the molar ratio of Mg/Al of 5.5 which is just within the appellants' lower limit of 5 and the ratio of 4.4 which is just below the appellants' conditions giving a 99% aluminum removal efficiency while the 4.4 Mg/Al molar ratio, other conditions being the same, gave a 79% Al removal efficiency.

It is believed that the foregoing comments and the discussion of the evidence of record show that the Examiner's rejection is in error and should be reversed, particularly since there is no suggestion in Nagy to use the appellants' specific combination of conditions or that such combination would give the highly effective Al removal the appellants have shown they obtain.

Summary

In summary, the appellants submit, for the reasons noted, that their method is new and not obvious from Nagy. The declaration evidence supports the unobviousness of the appellants' claimed combination of features in efficiently removing Al from brine solutions. The Examiner's Section 103(a) rejection should be reversed for the reasons given.

Respectfully submitted,

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APPENDIX

1. A method for the reduction of soluble aluminum species in an evaporated salt alkali metal halide brine containing up to 500 ppb aluminum species to provide a brine feedstock suitable for use in a chlor-alkali membrane cell process, said method comprsising:

(a) treating said brine with a magnesium salt in an amount to provide a Mg to Al molar ratio selected from 5-20 to 1 and at a Mg concentration of from 0.5 to 5 ppm, and sufficient alkali metal hydroxide to provide an excess alkalinity concentration of between 0.1-0.3 g/L alkali metal hydroxide to effect precipitation of a magnesium aluminum hydroxide complex; and

(b) removing said complex to provide said brine feedstock.

2. A method as defined in claim 1 wherein said Mg to Al molar ratio is about 10:1 and said Mg concentration is from 1 to 5 ppm.

3. A method as defined in claim 1 wherein said brine, said magnesium salt and said alkali metal hydroxide are subjected to vigorous mixing.

4. A method as defined in claim 1 wherein said brine is treated with said magnesium salt prior to treatment with said alkali metal hydroxide.

5. A method as defined in claim 1 wherein said magnesium salt and said alkali metal hydroxide are added to said brine as aqueous solutions.

6. A method as defined in claim 1 wherein said magnesium salt is magnesium chloride and said alkali metal is sodium.

7. A method as defined in claim 1 wherein said treating is carried out at 50-70°C and the excess alkalinity is between 0.1 to 0.2 g/L alkali metal hydroxide and

further comprising determining the concentration of aluminum species in said feedstock and adding magnesium salt to provide said Mg to Al ratio within said Mg concentration.

9. In a chlor-alkali process employing a brine feedstock, the improvement wherein said feedstock is obtained by the method of claim 1.

10. A chlor-alkali process according to claim 9 which is closed loop.